Quasi-Elastic Neutron Scattering from Methanol Associates

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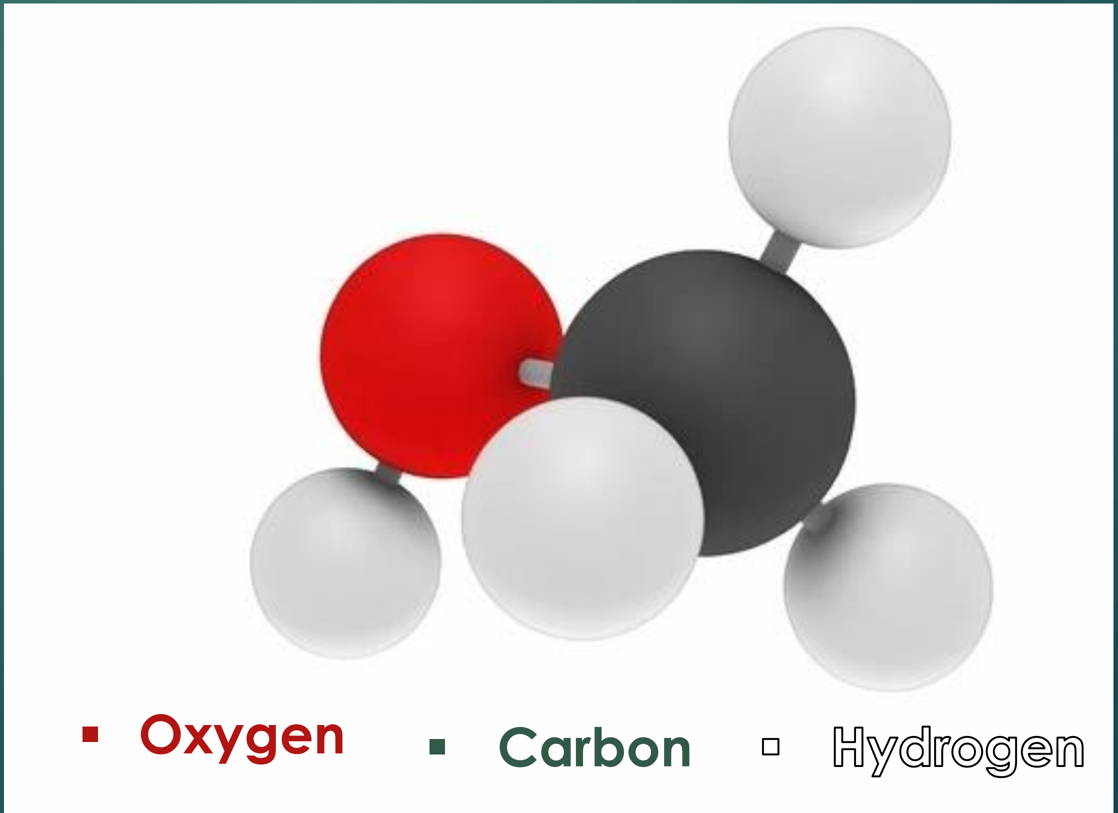
Overview

- Motivation
- Static Structuring of Methanol Associates
  - Observation of pre-peak
- Dynamics of Methanol Associates
  - Length-scale and temperature dependence
- Conclusion
- Acknowledgements

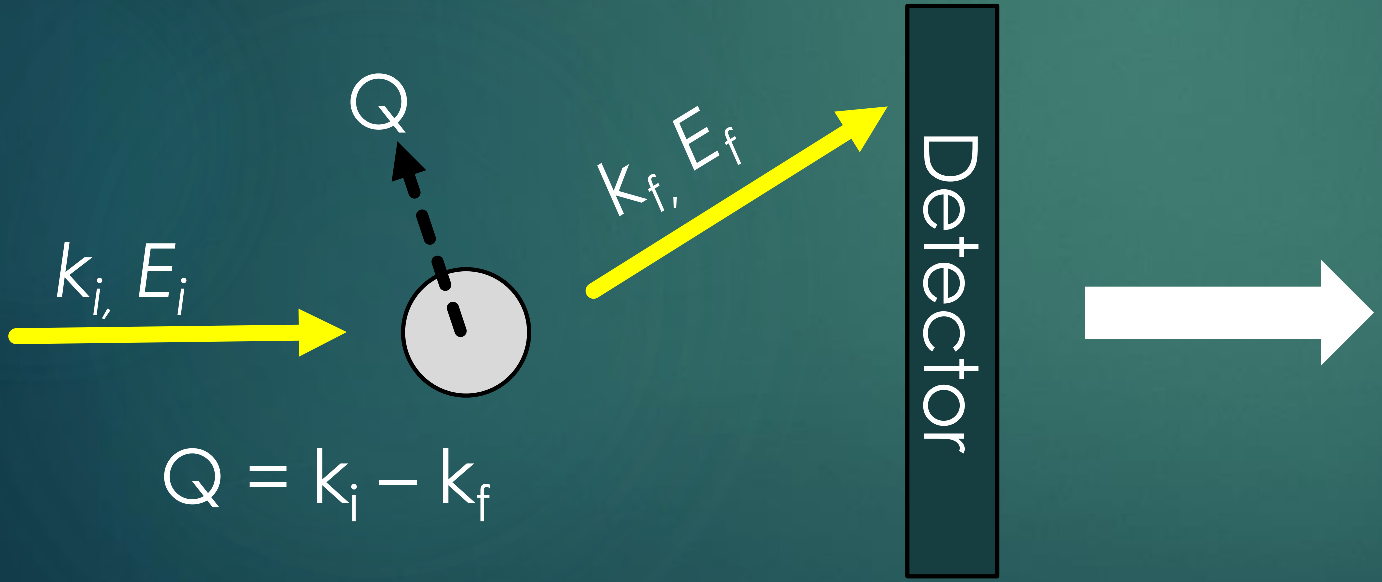
Why Methanol?

- Most fundamental amphiphile
- Static structuring is still debated
- Dynamic structuring has not been studied extensively



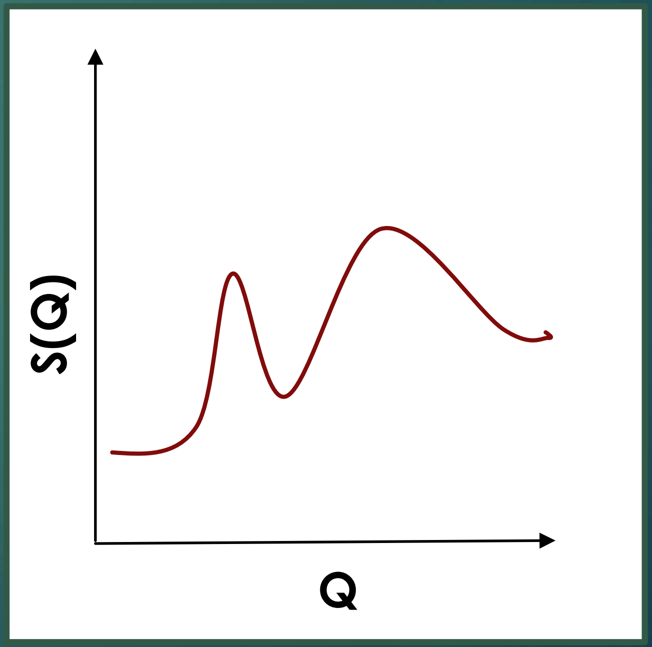
Neutron Scattering Fundamentals

- Neutrons interact with material by scattering off nucleus
- Detector records intensity and position



$Q = k_i - k_f$

$(E = E_i - E_f)$



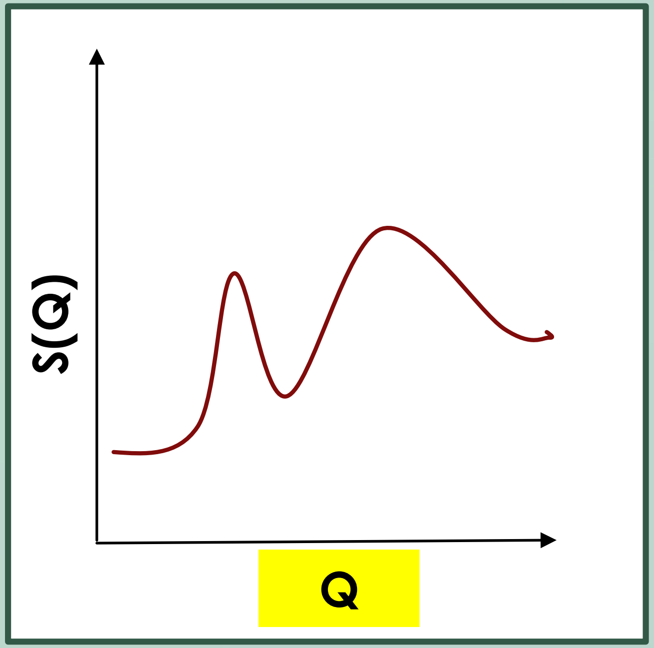
Scattering Function

Neutron Scattering Fundamentals

- Neutrons interact with material and are scattered by the strong nuclear forces
- Detector records intensity

Momentum Transfer (Q)

- Describes change in momentum of incident-to-resultant neutron
- Corresponds to the **inverse** of the length-scale probed
- Expressed in inverse Angstroms ($Å^{-1}$)

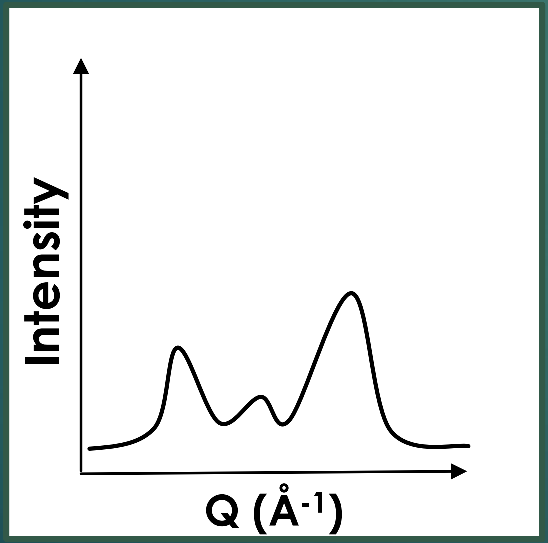


Scattering Function

Neutron Scattering Concepts

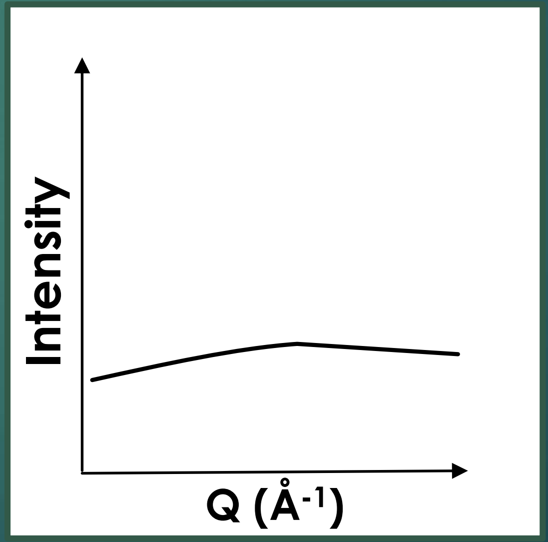
Coherent Scattering

- Contains information on correlation lengths and motions of atoms



Incoherent Scattering

- Contains information on the dynamics of single atoms



Isotopic Scattering

- Isotopes of the same element can have characteristically different scattering
  - Doesn’t change behavior of molecules, significantly

Hydrogen (H)

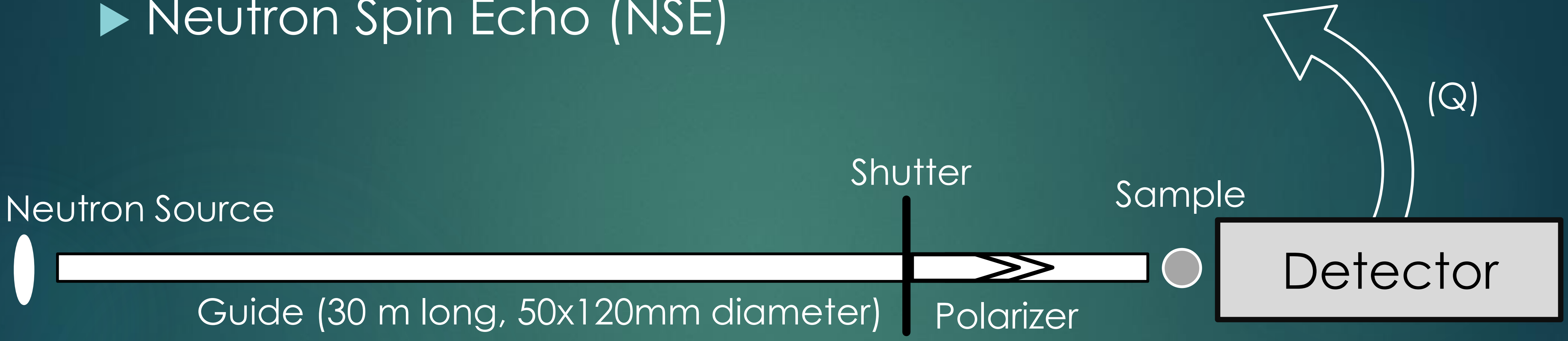
- Coherent = -3.74fm (13%)
- Incoherent = 25.27fm (87%)

Deuterium (D)

- Coherent = 6.67fm (62%)
- Incoherent = 4.04fm (38%)

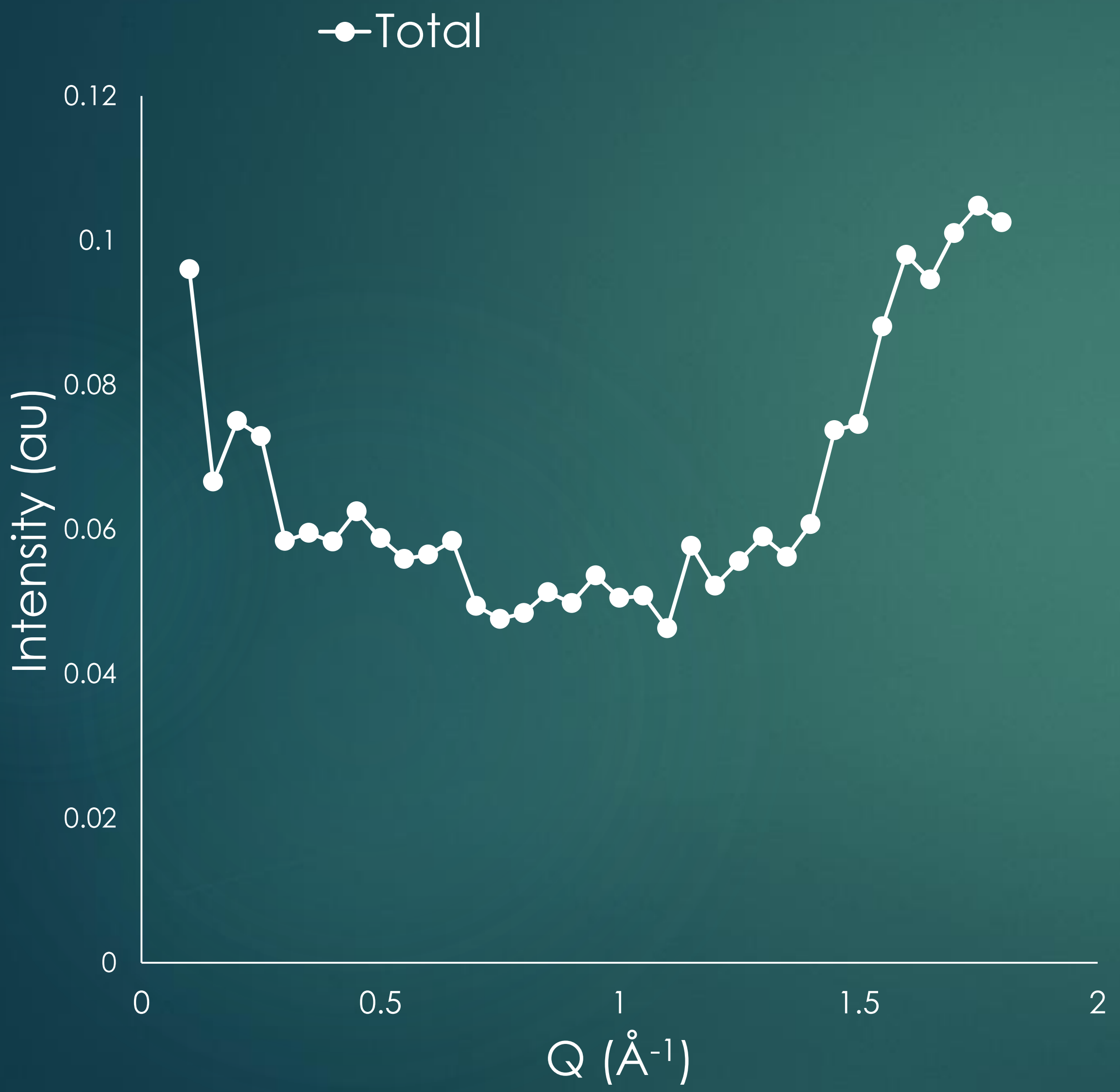
Polarized Diffraction

- Neutron Spin Echo (NSE)



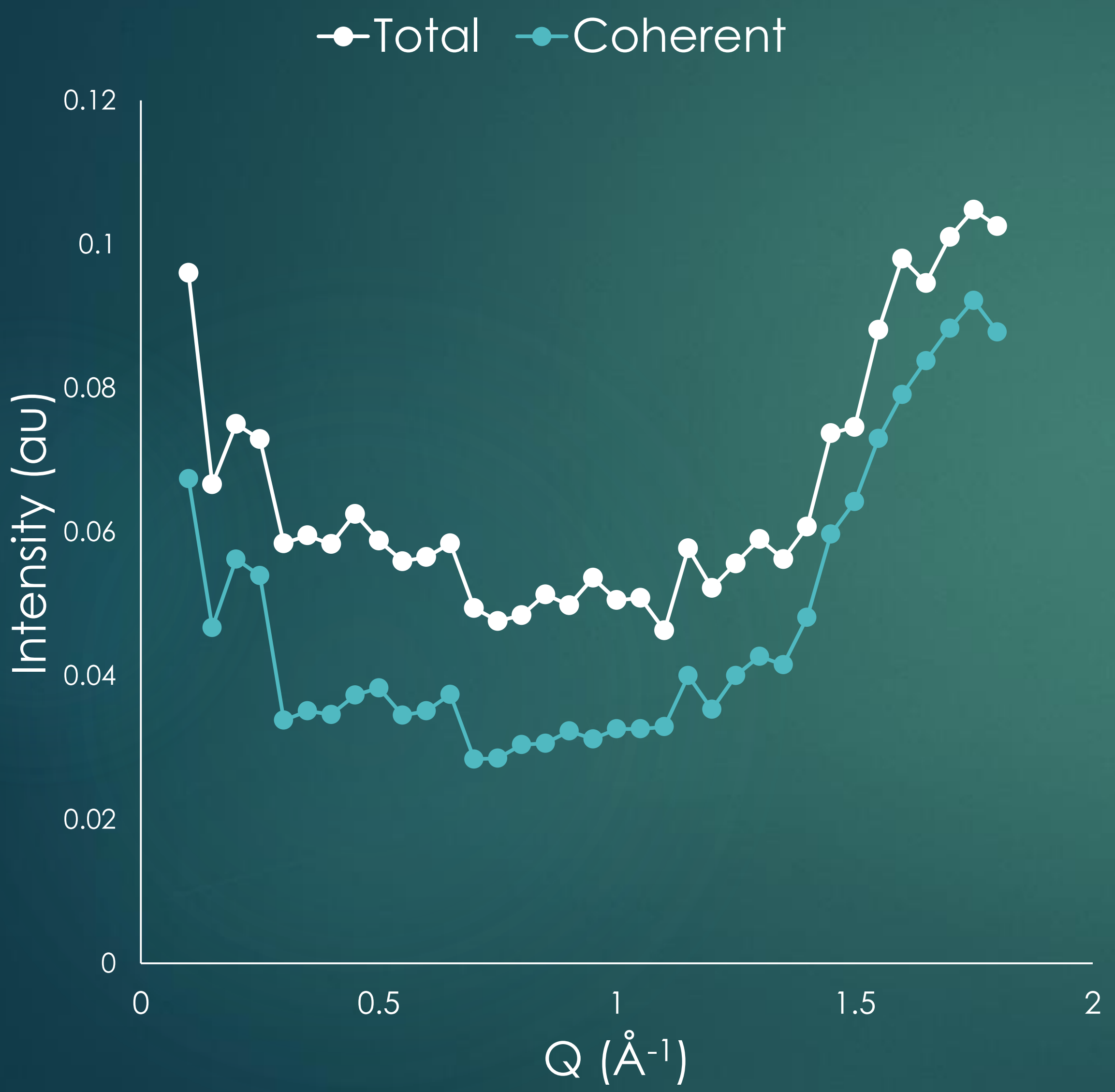
$$S_{Total}(Q) \rightarrow S_{Coherent}(Q) \,\&\, S_{Incoherent}(Q)$$

NSE Scattering Results



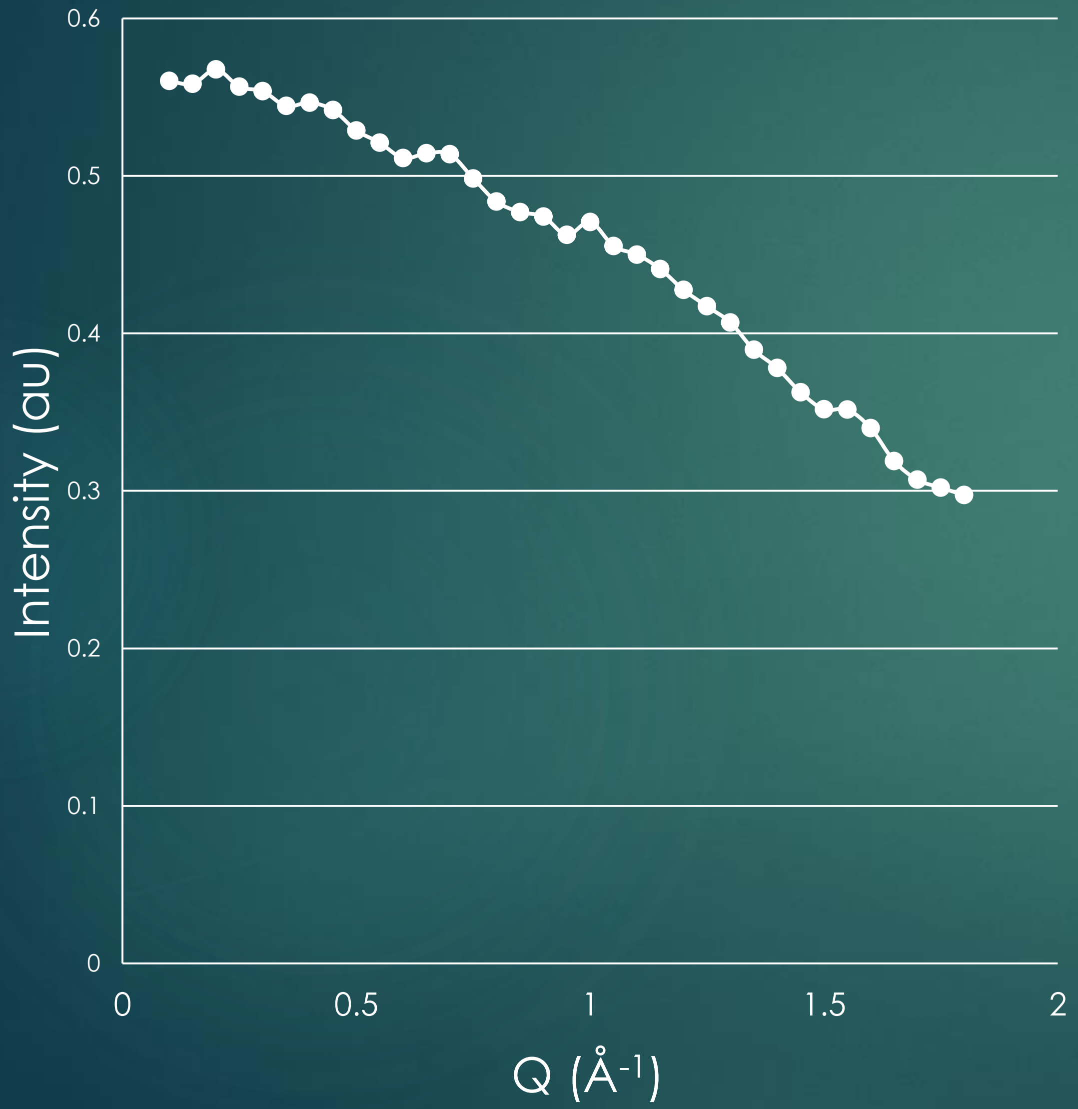
- CD_3OD Total Scattering
  - Structure peak at ≈1.80 Å^{-1}

NSE Scattering Results



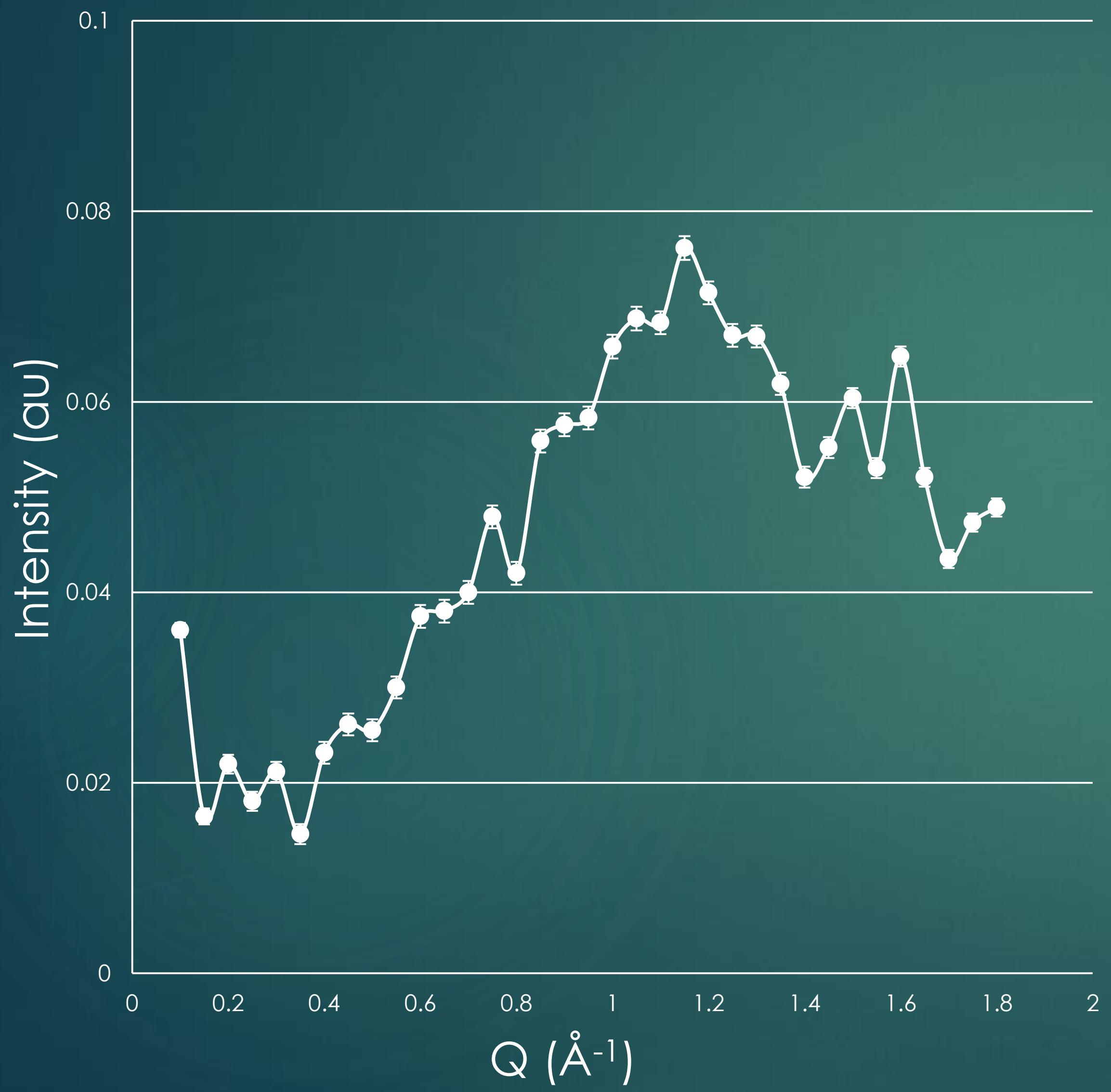
- CD_3OD Total Scattering
  - Structure peak at ≈1.80 $Å^{-1}$
- CD_3OD Coherent Scattering
  - Polarization allows for separation of incoherent and coherent data
  - Only slight difference because deuterium has small incoherent contribution

NSE Scattering Results



- CH_3OD Total Scattering
  - Incoherent noise from Hydrogen washes out key structural data

NSE Scattering Results

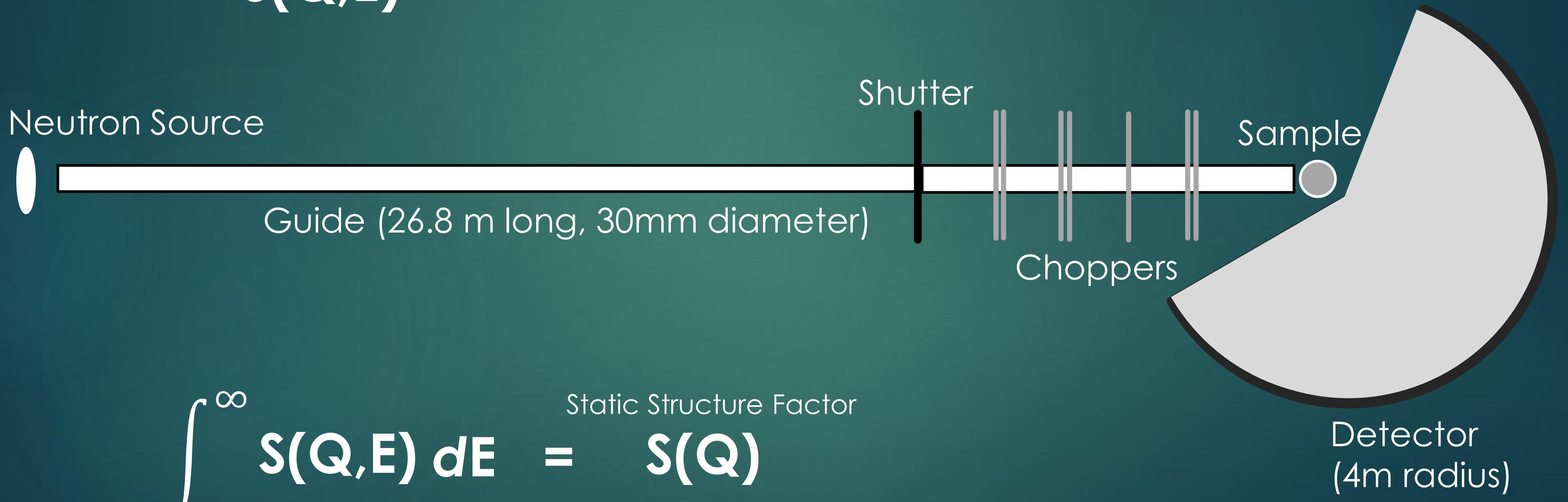


- CH_3OD Coherent Scattering
  - Polarization of incident neutron beam allows for separation of coherent and incoherent scattering
- Observation of new peak
  - "Pre-peak" at ≈1.10 $Å^{-1}$
  - Lower Q value indicates structuring at larger length-scales (associates)

Disk Chopper Spectrometer (DCS)

Dynamic Structure Factor

S(Q,E)



$$\int_{-\infty}^{\infty} S(Q,E)\, dE = S(Q)$$

Static Structure Factor

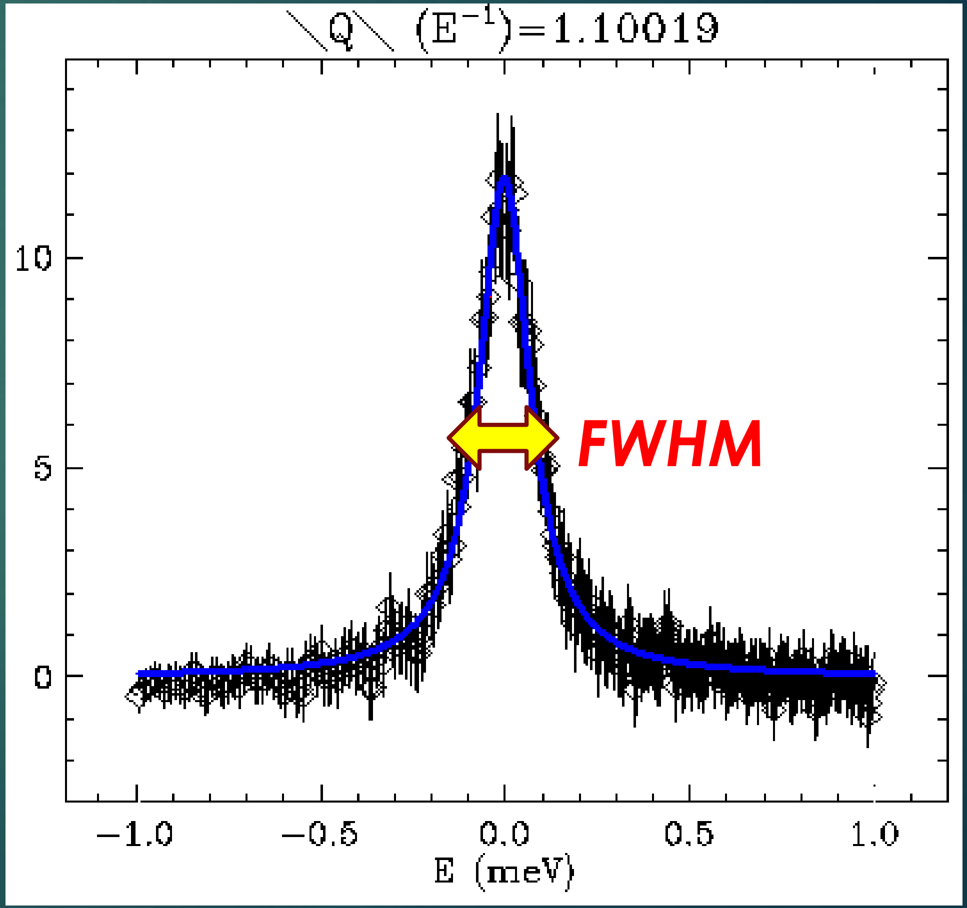
Exploring Dynamics

- Dynamic Scattering Function Model

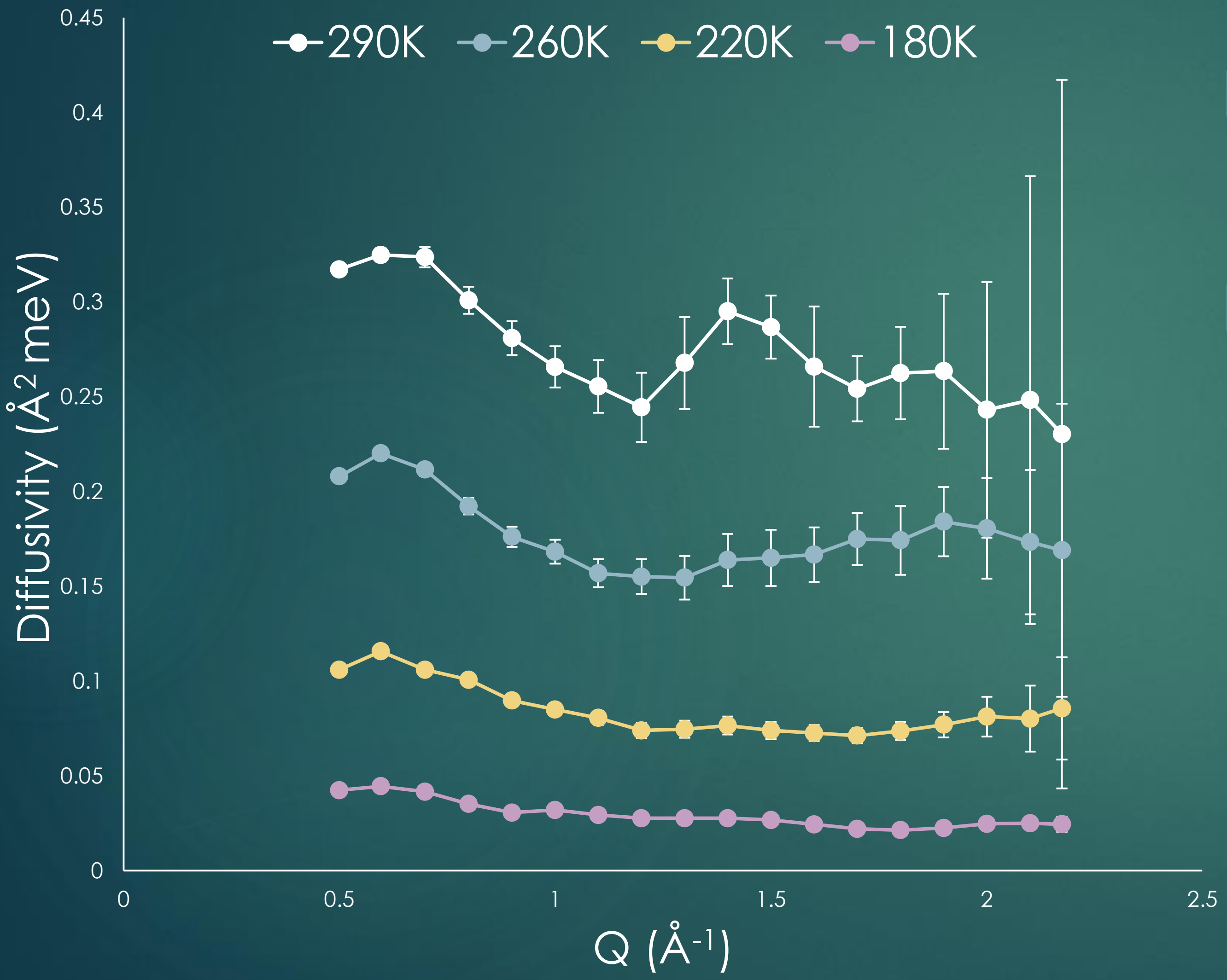
$$S(Q,E) = \frac{2A}{\pi}\left[\frac{FWHM}{4E^2+(FWHM)^2}\right] \otimes R(Q,E)$$

  - Dynamic data fitted with Lorentzian

- $(FWHM)$ is the main parameter
  - Quantifies time-scale of interaction



Dynamics of Structures



- CD_3OD Dynamics
- Effective Diffusion Constant:

$$D_{eff} = \frac{FWHM}{Q^2}$$

Data Combination

$$S^{Combination} = [S^{CH_3OD} + S^{CD_3OH}] - [S^{CD_3OD} + S^{CH_3OH}]$$

$$S^{Combination} =$$

$$[\ (b_{in}^{C}S_{in}^{C}+b_{in}^{D}S_{in}^{H_H} + b_{in}^{H}S_{in}^{H_m} + b_{in}^{O}S_{in}^{O}) + (b_{Co}^{C}b_{co}^{D}S_{Co}^{CH_H}+b_{Co}^{C}b_{co}^{H}S_{Co}^{CH_m} + b_{Co}^{C}b_{co}^{O}S_{Co}^{CO}+b_{Co}^{O}b_{co}^{D}S_{Co}^{CH_H}+b_{Co}^{O}b_{co}^{H}S_{Co}^{OH_m} + b_{co}^{D}b_{co}^{H}S_{Co}^{H_H H_m})$$

$$+ (b_{in}^{C}S_{in}^{C} + b_{in}^{D}S_{in}^{H_m} + b_{in}^{H}S_{in}^{H_H} + b_{in}^{O}S_{in}^{O}) + (b_{Co}^{C}b_{co}^{H}S_{Co}^{CH_H}+b_{Co}^{C}b_{co}^{D}S_{Co}^{CH_m} + b_{Co}^{C}b_{co}^{O}S_{Co}^{CO}+b_{Co}^{O}b_{co}^{H}S_{Co}^{CH_H}+b_{Co}^{O}b_{co}^{D}S_{Co}^{OH_m} + b_{co}^{D}b_{co}^{H}S_{Co}^{H_H H_m})]$$

$$-[\ (b_{in}^{C}S_{in}^{C} + b_{in}^{D}S_{in}^{H_m} + b_{in}^{D}S_{in}^{H_H} + b_{in}^{O}S_{in}^{O}) + \left(b_{Co}^{C}b_{co}^{D}S_{Co}^{CH_H}+b_{Co}^{C}b_{co}^{D}S_{Co}^{CH_m} + b_{Co}^{C}b_{co}^{O}S_{Co}^{CO}+b_{Co}^{O}b_{co}^{D}S_{Co}^{CH_H}+b_{Co}^{O}b_{co}^{D}S_{Co}^{OH_m} + b_{co}^{D}b_{co}^{D}S_{Co}^{H_H H_m}\right)$$

$$+ (b_{in}^{C}S_{in}^{C} + b_{in}^{D}S_{in}^{H_m} + b_{in}^{H}S_{in}^{H_H} + b_{in}^{O}S_{in}^{O}) + (b_{Co}^{C}b_{co}^{H}S_{Co}^{CH_H}+b_{Co}^{C}b_{co}^{H}S_{Co}^{CH_m} + b_{Co}^{C}b_{co}^{O}S_{Co}^{CO}+b_{Co}^{O}b_{co}^{H}S_{Co}^{CH_H}+b_{Co}^{O}b_{co}^{H}S_{Co}^{OH_m} + b_{co}^{H}b_{co}^{H}S_{Co}^{H_H H_m})]$$

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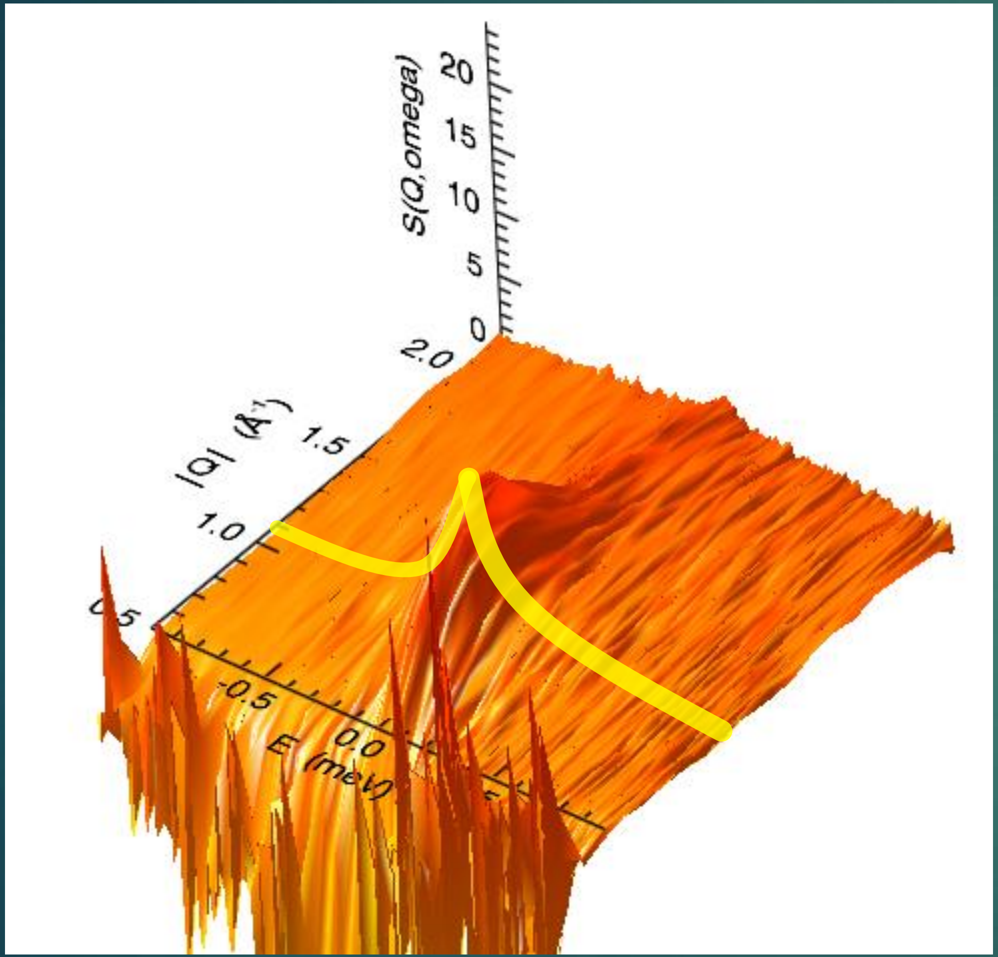
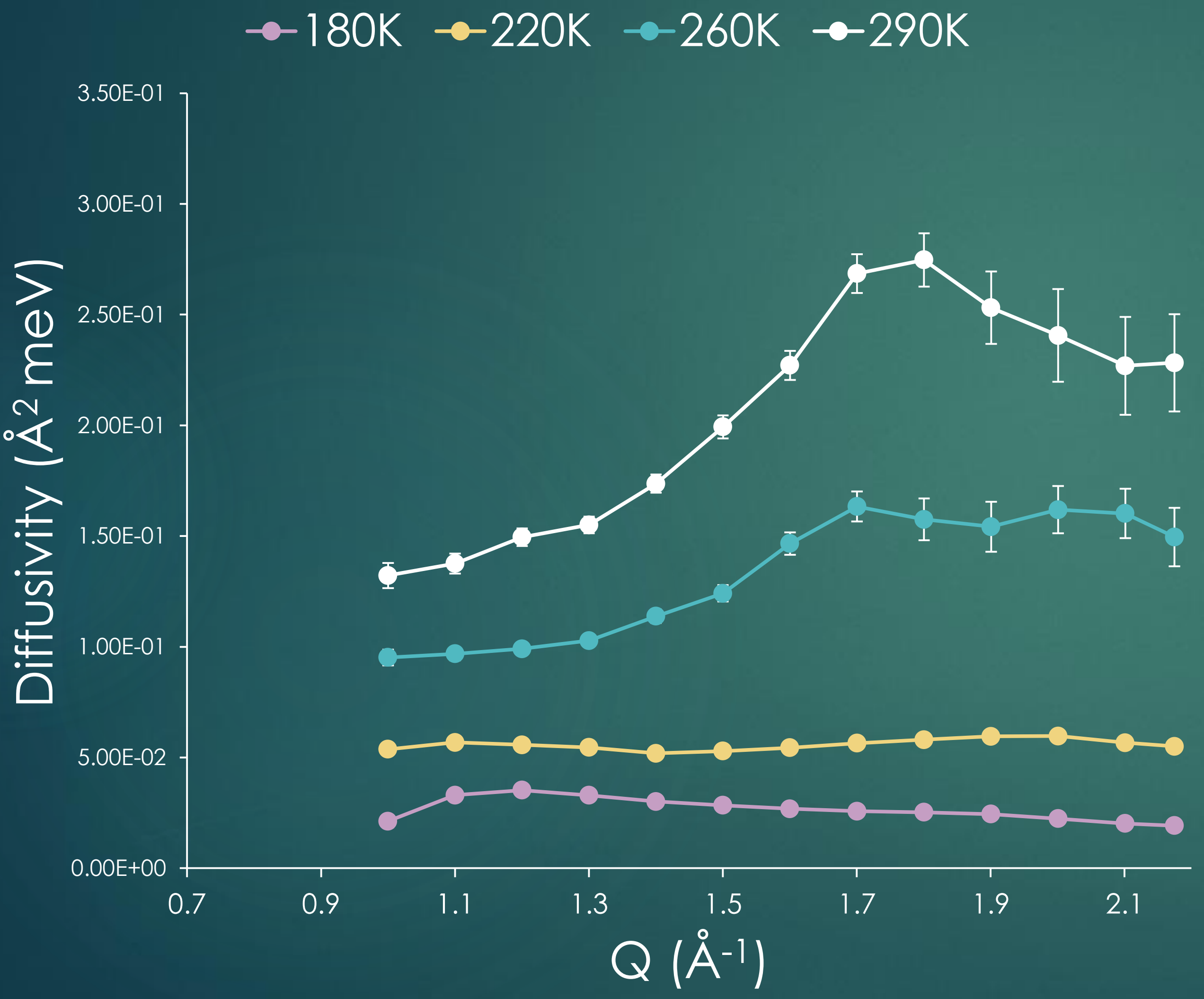
Eliminates incoherence, isolates Methyl-Hydroxyl Interaction

DCS Scattering Results



- Combination isolates specific interaction:
  - Methyl-Hydroxyl
- Pre-peak observed at ≈1.10 Å^{-1}

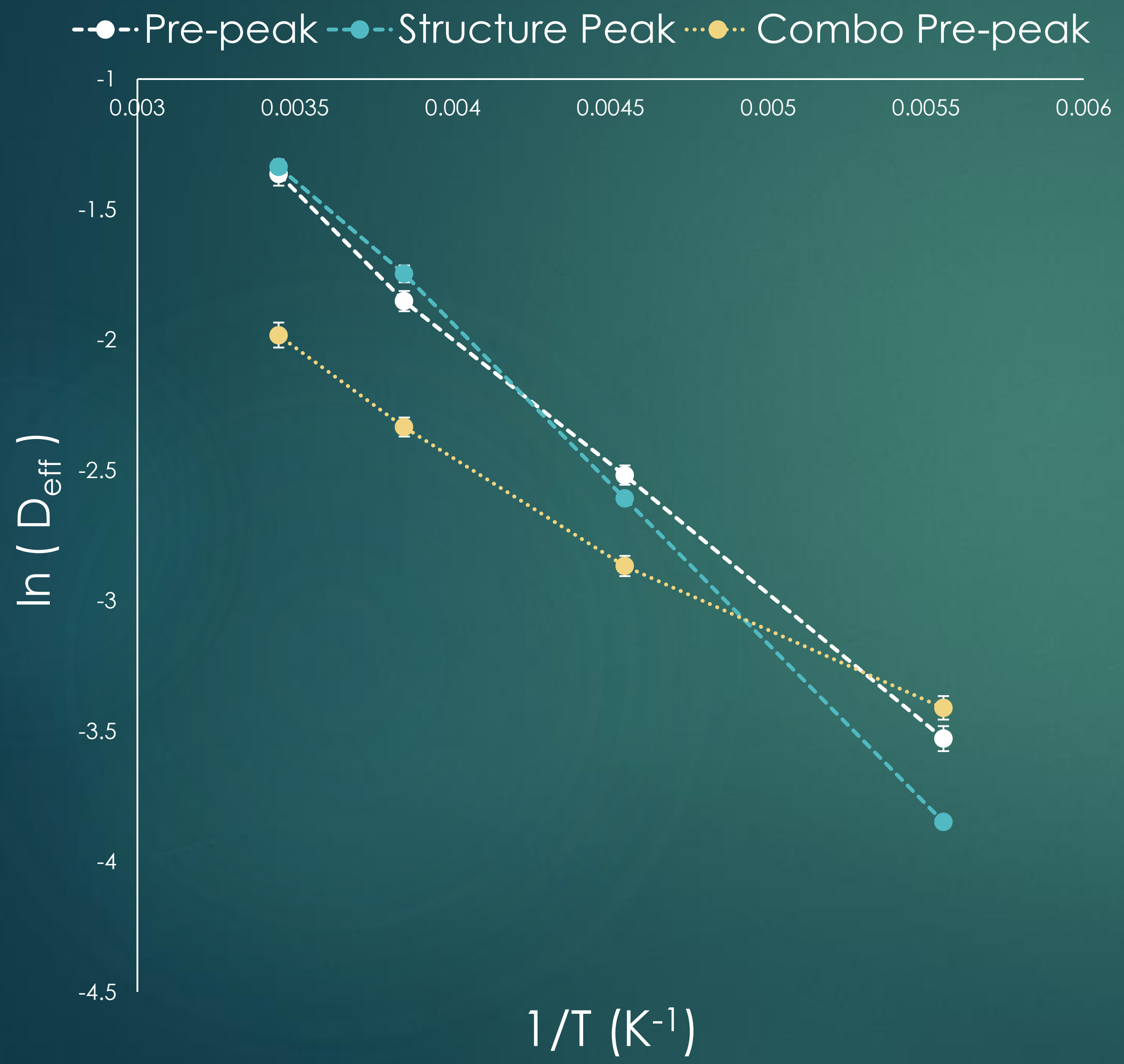
Dynamics of Structures



- Combination Dynamics
- Effective Diffusion Constant:

$$D_{eff} = \frac{FWHM}{Q^2}$$

Dynamics of Structures



- Arrhenius Plot
  - Linearity shows direct temperature dependence of interactions
  - Can calculate approximate activation energy of interactions

Activation Energies:

Structure peak

E_A = 9.988 kJ mol^{-1}

Pre-peak

E_A = 8.427 kJ mol^{-1}

Conclusion

- Further characterized a Methanol pre-peak, using neutron scattering
  - Pre-peak observed at ≈1.10 $Å^{-1}$
- Began investigating the dynamic behavior of this structure

Acknowledgements

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